EPJ Web of Conferences **146**, 11022 (2017) DOI: 10.1051/epjconf/201714611022
*ND2016*

# Measurement of the $^{241}$Am neutron capture cross section at the n_TOF facility at CERN

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**Abstract.** New neutron cross section measurements of minor actinides have been performed recently in order to reduce the uncertainties in the evaluated data, which is important for the design of advanced nuclear reactors and, in particular, for determining their performance in the transmutation of nuclear waste. We have measured the $^{241}$Am(n,$\gamma$) cross section at the n_TOF facility between 0.2 eV and 10 keV with a $BaF_2$ Total Absorption Calorimeter, and the analysis of the measurement has been recently concluded. Our results are in reasonable agreement below 20 eV with the ones published by C. Lampoudis et al. in 2013, who reported a 22% larger capture cross section up to 110 eV compared to experimental and evaluated data published before. Our results also indicate that the $^{241}$Am(n,$\gamma$) cross section is underestimated in the present evaluated libraries between 20 eV and 2 keV by 25%, on average, and up to 35% for certain evaluations and energy ranges.

## 1. Introduction

Isotopes of $^{241}$Am and $^{237}$Np are the minor actinides which contribute most to the long term hazard of the nuclear waste. The first one, with $T_{1/2}$ = 433 y, decays into $^{237}$Np ($T_{1/2} = 2.1 \times 10^6$y) via $\alpha$ emission. Neutron capture reactions in $^{241}$Am, the most abundant Am isotope present in the spent nuclear fuel, are the path to heavier minor actinides like $^{243}$Am and Cm isotopes. In addition, the reprocessing of Am is technologically more advanced than for other minor actinides. For all these reasons the improvement of the $^{241}$Am(n,$\gamma$) data was a major effort of the EC-FP7 ANDES project, and four different measurements were planned and carried out. Two of them were capture measurements performed at n_TOF [1], with the same sample but different detectors: the Total Absorption Calorimeter (TAC) [2] and $C_6D_6$ detectors [3]. The other two were a transmission and a capture measurement performed at IRMM – Geel [5], with a different sample. In this work we present the results of the measurement performed at n_TOF with the TAC.

## 2. Experimental setup

The measurement was performed in 2010 at the n_TOF facility at CERN, which is a high instantaneous intensity spallation neutron source driven by the CERN PS synchrotron. We used the Total Absorption Calorimeter (TAC) [2] to detect the capture reactions as a function of the time-of-flight. This detector is made with 40 $BaF_2$ crystals of 15 cm length covering $\sim$95% of the solid angle, and is used to detect in coincidence (nearly) all the $\gamma$-rays coming from the (n,$\gamma$) reactions. The neutron beam was monitored during the entire measurement with a $^6$Li foil based detector [6]. The detector signals were recorded by a digital data acquisition system [7] operating at 250 and 500 MSamples/s with 8 bits resolution, recording continuously a time of flight of 32 or 16 ms for each pulse (above 0.17 or 0.7 eV, since the time of flight is about 185 m).

The sample [8] was made of 36.5 mg of $^{241}AmO_2$ embedded in a 305 mg $Al_2O_3$ matrix (to reduce/avoid inhomogeneities), and encapsulated in a 0.5 mm thick Al canning. The radius of the sample was 6.13 mm and the $^{241}$Am mass 32.23(10) mg ($6.82 \cdot 10^{-5}$ atoms/barn). It was placed in the center of the TAC, shielded with 2 mm lead due to its high activity ($\sim$4 GBq), and a spherical borated polyethylene neutron absorber (5/10 cm inner/outer radius), used to reduce the background due to scattered neutrons. In the analysis of the measurement we observed resonances of $^{237}$Np and $^{240}$Pu. We estimated the amount of these impurities as $\sim$0.029 ($^{237}$Np) and $\sim$0.00049 ($^{240}$Pu) times the number of $^{241}$Am atoms.

About 2 months of beam time were dedicated to the $^{241}$Am measurement. 60% of the total amount of beam was dedicated to measure the $^{241}$Am sample, 36% to measure the different background components, and 4% to measure different $^{197}$Au samples in order to normalize the $^{241}$Am(n,$\gamma$) cross section by means of the saturated resonance method [9], using the $^{197}$Au strongest resonance at 4.9 eV. The number of events detected per proton pulse during the $^{241}$Am measurement and some of the background measurements (a dummy sample and the Al canning, and measurements without neutron beam with and without the sample in place) are presented in Fig. 1. Only the events with certain conditions in the detection multiplicity $m_{cr}$ (number of detectors in coincidence) and total deposited energy $E_{sum}$ (sum of the energies deposited in the detectors in coincidence) have been considered. Sm impurities present in the $Al_2O_3$ matrix of the dummy sample but not in the $^{241}$Am sample are clearly visible.

## 3. Data analysis

The basis of the analysis procedure was the same as other TAC measurements [10,11], with some additional features developed to deal with pulse pile-up effects not present in previous analysis.

The detection efficiency has been determined from simulations, based in the DECAYGEN [12] code for the generation of the (n,$\gamma$) cascades and in a very detailed geometric modeling of the TAC [13] implemented in GEANT4 [14] for the transport of the $\gamma$-rays. The Photon Strength Functions of the compound nuclei ($^{242}$Am and $^{198}$Au) used by DECAYGEN were modified until reproducing the experimental results (deposited energy histograms for different detection multiplicities), as it has been done in previous works [10,11]. An example of how well the experimentally obtained deposited energy distributions due to $^{241}$Am(n,$\gamma$) cascades are reproduced by the simulations is presented in Fig. 2.

Three different $^{197}$Au samples were measured to normalize to the saturated resonance at 4.9 eV. The counting rates in the saturated part of the resonance

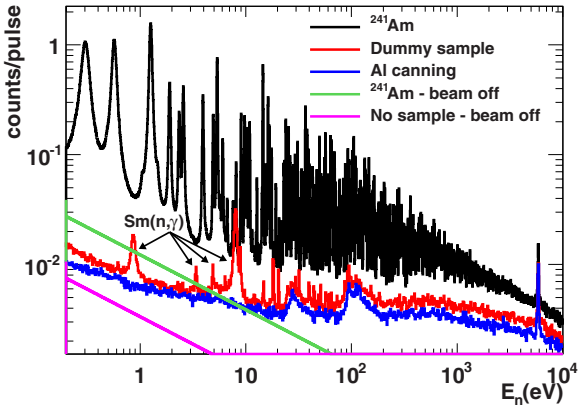


**Figure 1.** Number of events detected in the $^{241}$Am(n,$\gamma$) measurement as a function of the neutron energy, together with different background measurements and under the conditions of $m_{cr} > 2$ and $2.5 < E_{sum} < 6$ MeV.

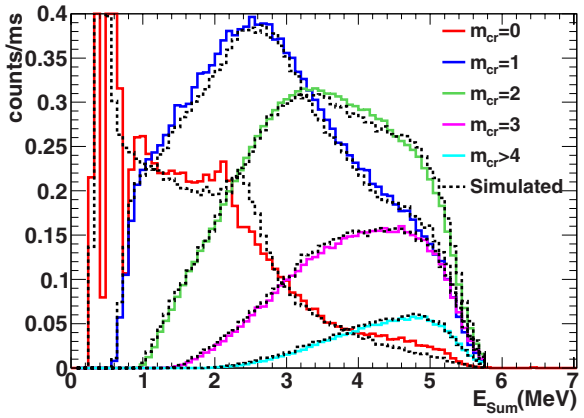


**Figure 2.** Experimental (solid lines) and simulated (dotted lines) deposited energy spectra from $^{241}$Am capture cascades for different $m_{cr}$ conditions.

were quite high for the TAC (a few events per $\mu$s), and we used the third method described in [15] to correct for the pile-up effects. Each of the measurements were performed at two different pulse intensities, being the highest about 2.3 times more intense than the lowest. All the normalization values obtained with the different samples and the different pulse intensities were compatible within statistical uncertainties, which were close to 1% in all the cases. The overall normalization uncertainty is the sum of the uncertainties of the sample mass (0.6%), the detection efficiency (2%) and normalization to the $^{197}$Au resonance (2%), which amounts to 3% or 5%, depending on whether the quantities are added quadratically or linearly.

The background components not related with the interaction of the neutron beam with the $^{241}$Am nuclei and the $^{237}$Np and $^{240}$Pu impurities were obtained from dedicated measurements. The background due to fission reactions in $^{241}$Am is very low, and was estimated from the JEFF-3.2 [16] fission cross section and by assuming a similar detection efficiency than for the capture reactions. The background due to elastic scattered neutrons in $^{241}$Am was estimated by measuring the effect of neutrons scattered in a graphite sample together with the JEFF-3.2 $^{241}$Am elastic cross section. The effect of this component is negligible in the Resolved Resonance Region (RRR), since the neutron absorber is more effective at lower neutron energies, and very low ($\sim$ 1%) in the Unresolved Resonance Region (URR). The effect of the $^{237}$Np and the $^{240}$Pu impurities were also taken into account from the cross section data available in the evaluations.

The self-shielding and multiple scattering effects due to the $^{241}$Am sample and to the Al canning and the $Al_2O_3$ matrix have been carefully studied by comparing calculations performed with SAMMY [17], MCNP-6.1 [18] and GEANT4. In these calculations we obtained the theoretical capture yield under different conditions (with/without the Al canning, the $Al_2O_3$ matrix, ...), finding that the results obtained with the three codes were in a reasonable agreement. The effect due to the $^{241}$Am is small (<2%) except for the first three resonances, where is around 10%. The effect of the Al canning is negligible, and the effect of the $Al_2O_3$ matrix is to increase the number of detected capture events by around a 2%, nearly constant in the energy range of interest.

Above 1–2 keV there are pile-up effects in the capture signals induced by the high background counting rate. This effect has been corrected with a method which is quite similar to the ones described in [15] and that will be published in a future work. Below 1–2 keV this effect is very small.

The obtained capture yield has been analyzed with SAMMY, using a resonant description of the cross section up to 700 eV. The URR was analyzed up to 10 keV with the modified version of the FITACS code available in SAMMY.

## 4. Results

Together with this measurement, other three $^{241}$Am(n,$\gamma$) measurements have been performed recently: one at n_TOF using the same sample but $C_6D_6$ detectors instead of the TAC [3] (Fraval et al.); other (together with transmission) at IRMM – Geel [5] (Lampoudis et al.) using a three times thicker sample, manufactured in the same laboratory and at the same time as the one used in this work; and the last one performed at DANCE [4] (Jandel et al.), in Los Alamos Neutron Science Center.

Lampoudis et al. provided a capture cross section up to 110 eV which was, on average, around 20% larger than the results provided by Fraval et al. and Jandel et al., which are in reasonable agreement between them. The JEFF-3.2 neutron data library has adopted the results provided by Lampoudis et al. in the RRR (up to 150 eV) whereas the JENDL-4.0 [19] and ENDF/B-VII.1 [20], which are the same in the RRR, have adopted the results provided by Jandel et al. In the URR the three evaluated data libraries are quite similar.

A comparison between the $^{241}$Am(n,$\gamma$) cross section we obtained and the evaluated ones is presented in Fig. 3. Our results are a bit below JEFF-3.2 for the first three resonances, and in in reasonable agreement up to 20 eV. Between 20 eV and 150 eV, our cross section is higher than the one in JEFF-3.2 by up to 10–15%, and also higher than the one in JENDL-4.0 and ENDF/B-VII.1 by up to 30–35%. In the low energy part of the URR, our results are higher than the evaluated ones by up to 20%, but above 1–2 keV our $^{241}$Am(n,$\gamma$) cross section is compatible with the evaluated ones.

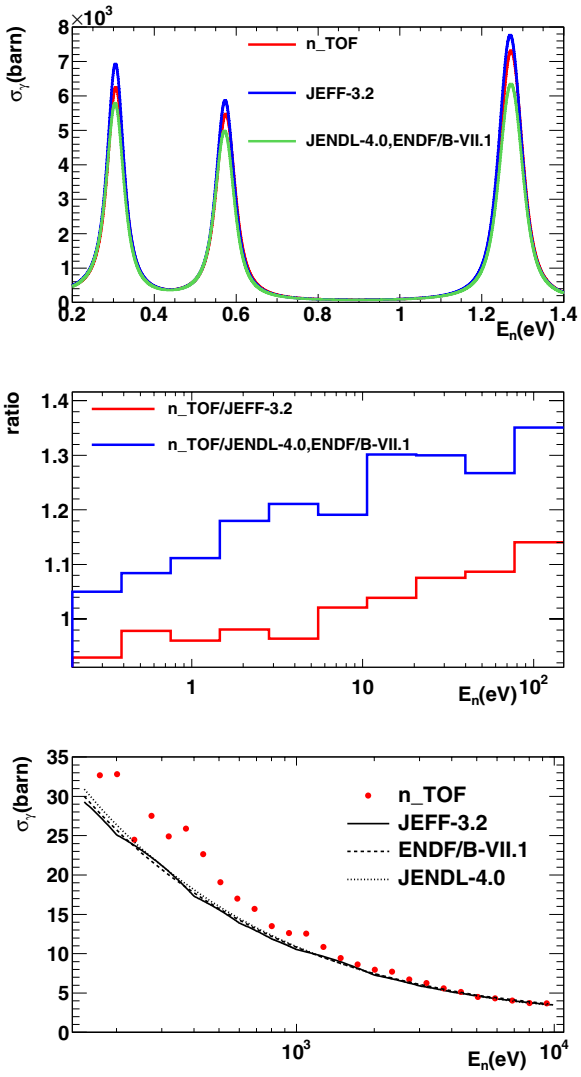


**Figure 3.** Comparison between the obtained cross section and the evaluated ones. In the top pannel, the different cross sections for the first three $^{241}$Am resonances. In the middle, the ratio between them in the RRR. In the bottom panel, the different cross sections in the URR up to 10 keV.

## 5. Conclusions

We have measured the $^{241}$Am(n,$\gamma$) cross section at n_TOF with the TAC between 0.2 eV and 10 keV. Our results are compatible with the ones provided by Lampoudis et al. below 20 eV and with the evaluated cross sections above 1–2 keV. This results indicate that the present evaluations underestimate the $^{241}$Am(n,$\gamma$) cross section below 1 keV by up to 30%.

This work was supported in part by the Spanish national company for radioactive waste management ENRESA, through the CIEMAT-ENRESA agreements on "Transmutación de residuos radiactivos de alta actividad" the Spanish Plan Nacional de I+D+i de Física de Partículas (project FPA2014-53290-C2-1) and the European Commission with the FP7 projects ANDES (FP7-249671) and CHANDA (FP7-605203).

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